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Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions

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Abstract

A large number of published and unpublished measurements of cloud condensation nuclei (CCN) concentrations and aerosol optical thickness (AOT) measurements have been analyzed. AOT measurements were obtained mostly from the AERONET network, and selected to be collocated as closely as possible to the CCN investigations. In remote marine regions, $CCN_{0.4}$ (CCN at a supersaturation of 0.4%) are around 110 cm^{-3} and the mean AOT_{500} (AOT at 500 nm) is 0.057. Over remote continental areas, CCN are almost twice as abundant, while the mean AOT_{500} is ca. 0.075. (Sites dominated by desert dust plumes were excluded from this analysis.) Some, or maybe even most of this difference must be because even remote continental sites are in closer proximity to pollution sources than remote marine sites. This suggests that the difference between marine and continental levels must have been smaller before the advent of anthropogenic pollution.

Over polluted marine and continental regions, the CCN concentrations are about one magnitude higher than over their remote counterparts, while AOT is about five times higher over polluted than over clean regions. The average CCN concentrations from all studies show a remarkable correlation to the corresponding AOT values, which can be expressed as a power law. This can be very useful for the parameterization of CCN concentrations in modeling studies, as it provides an easily measured proxy for this variable, which is difficult to measure directly. It also implies that, at least at large scales, the radiative and microphysical effects of aerosols on cloud physics are correlated and not free to vary independently. While this strong empirical correlation is remarkable, it must still be noted that that there is about a factor-of-four range of CCN concentrations at a given AOT, and that there remains considerable room for improvement in remote sensing techniques for measuring CCN abundance.

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1 Introduction

The concentration of cloud-active particles, especially in the lower troposphere, has a profound influence on the microphysical processes in clouds, and consequently on many aspects of weather and climate. These interactions have been summarized in a number of recent reviews, addressing in particular the effects of aerosols on climate (Penner et al., 2001; Lohmann and Feichter, 2005; IPCC, 2007) and on cloud processes and precipitation (McFiggans et al., 2006; Rosenfeld, 2006a; IAPSAG, 2007; Andreae and Rosenfeld, 2008). In addition to their cloud microphysical effects, aerosols also modulate cloud formation and convective behavior through their radiative effects, for which aerosol optical thickness (AOT) is a commonly used metric. AOT has the advantage of being readily observed by remote sensing, and AOT measurements are now done routinely from space by several sensors (Kaufman et al., 2002; Yu et al., 2003; Kahn et al., 2007; Kokhanovsky et al., 2007) as well as by ground-based sunphotometer networks (Holben et al., 2001; Kim et al., 2008).

In order to incorporate the effects of cloud condensation nuclei (CCN) in meteorological models at all scales, from large eddy simulation (LES) to global climate models (GCM), knowledge of the spatial and temporal distribution of CCN in the atmosphere is essential. This information is, however, difficult to obtain from observations. In-situ measurements of CCN concentrations only provide very localized and sparse information, while the detection of CCN by remote sensing has not yet been accomplished. This is due to the difference in size ranges important for CCN concentrations on one hand and for light extinction on the other.

The ability of a particle to nucleate a cloud droplet depends on its size and composition. The latter is now frequently represented by the hygroscopicity factor, κ, which typically falls in the range of 0.1–0.9 for ambient aerosols (Petters and Kreidenweis, 2007; Andreae and Rosenfeld, 2008). At these values of κ, particles must have diameters larger than about 40–70 nm in order to activate at the highest supersaturations commonly found in clouds (up to about 0.6%). This therefore represents the lower

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boundary of the size range containing CCN-active particles. The upper boundary is effectively defined by the sharp decrease in the number concentration of particles as a function of size, with the result that usually only a minor fraction of CCN is in the size range above some 200–300 nm diameter (Seinfeld and Pandis, 1998). The maximum of the CCN size distribution thus typically falls in the range of about 70–200 nm. This range is well below the maximum in the Mie scattering efficiency function for light with a wavelength of about 500 nm, which is most commonly used to represent the aerosol optical thickness (AOT) of the atmospheric column. In contrast, the maximum of the scattering and extinction efficiency functions often falls near or above the maximum of the mass size distribution of the aerosol in the range between 400 and 1000 nm, so that this part of the size distribution usually has the strongest influence on the AOT_{500} values. In regions with high loadings of dust and seasalt aerosol, the coarse mode (>1 μm diameter) may also contribute strongly to AOT_{500}.

The disconnect between the parts of the aerosol size spectrum dominating the CCN abundance and those dominating visible light extinction suggests that correlations between these two variables may not be very strong. Consequently, the use of remote sensing measurements for the estimation of CCN abundances has been considered difficult (Gasso and Hegg, 2003; Ghan et al., 2006; Kapustin et al., 2006; Rosenfeld, 2006b). However, because of the interest in using CCN concentrations and AOT values as parameters to represent the effects of aerosols in cloud models, and because a substantial amount of data for both variables has recently become available, I decided to make an empirical investigation of their correlation in collocated (or at least nearly collocated) data sets.

2 Methods

Most AOT values were obtained from the AERONET database publicly available on the Internet at http://aeronet.gsfc.nasa.gov/. These data have the advantage of being available from a global network of stations with consistent processing algorithms and

quality control (Dubovik et al., 2000). In most cases, I was able to use Level 2.0 data, which are fully calibrated and cloud-screened. In some instances, only Level 1.5 data were available, which do not yet have the final calibration applied. These cases, and the few cases where other sunphotometer data were used, have been indicated in the tables below. I calculated the statistics reported in the tables based on the daily average data reported in the AERONET database, in order to avoid introducing bias from variability in the number of measurements available on individual days. I chose not to use MODIS AOT data for this analysis because of persistent uncertainties regarding the absolute accuracy of this data (Li et al., 2007).

I chose to use AOT values at 500 nm (AOT_{500}), because they are most commonly available and most frequently used as a metric for aerosol burdens. When only measurements at other wavelengths were available, I chose the nearest available wavelength and made an adjustment using the appropriate Ångstrom exponent.

To represent CCN concentrations, I used the set of particles that activate at a supersaturation of 0.4%, a value commonly used for convective clouds. Many of the data were taken from the literature and have been obtained using a variety of instruments, about which details can be found in the original papers referenced in the tables. Our own previously unpublished measurements were obtained with the static chamber counter described by Frank et al. (2007) and the DMT continuous-flow counter using the techniques discussed in Rose et al. (2008). When only data at supersaturations other than 0.4% were available, they were adjusted to 0.4% using the conventional power law formulation for the dependence of CCN concentrations on supersaturation, S ($[CCN_S]=[CCN_{1.0}]\times S^k$). The exponent, k, was either derived from the data, or where this was not possible, I used default values of 0.5 for continental and 0.4 for marine sites.

In order to increase our database, I deduced CCN concentrations from measurements of aerosol size spectra for some cases where direct CCN measurements were not available. For this purpose, I used lower cutoff diameters specified in the Comment column in Table 2, chosen based either on measurements of these cutoff diameters

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from the same site or from similar locations, or obtained using appropriate values of κ, and the relationships between κ and the activation diameter given by Petters and Kreidenweis (2007). Obviously, this introduces additional uncertainty into our estimates, which is in each case a function of the accuracy of the estimate of the cutoff diameter and the shape of the size distribution. In unfavorable cases, this uncertainty may be large (up to about 50%). In most cases, however, the maximum of the size distributions at our rural and remote stations was well above the range of possible cutoff diameters (50–90 nm for the range of κ and supersaturations considered here), and therefore the resulting uncertainty is likely not greater than what one must accept when using literature data from a great variety of groups and instruments. Since we are looking only for fairly broad relationships and consider a wide range of datasets and conditions, it is unlikely that this approach would introduce a systematic bias.

In addition to data sets where corresponding pairs of AOT and CCN data were available, some values where only one of the variables was measured have been added to the tables for information purposes. Furthermore, condensation nuclei (CN) concentrations have been provided in the tables, when available. These data have been obtained by a variety of instruments, with different lower cut-off diameters, and thus some caution must be exercised when comparing their values.

3 Results and discussion

For our study, I have separated the available data into four general regimes: continental-remote, continental-polluted, marine-remote and marine-polluted. I have excluded the analysis of dust-dominated regions downwind of the major dust source regions from our analysis, because there is little or no collocated CCN and AOT data available, and because dominance of the coarse mode in these regions precludes a meaningful relationship between CCN and AOT.

An emphasis on looking at remote regions came from our interest in estimating the pre-human aerosol loading of the atmosphere, which is of relevance to understanding

the human perturbation (Andreae, 2007). At the present time, the most remote, and therefore probably closest to pristine continental regions are found in Amazonia and in parts of northern North America and Siberia. Because of the seasonal heavy pollution of the Amazonian atmosphere with biomass smoke, this area also provides the opportunity to study very clean and highly polluted conditions within the same region.

3.1 Amazonia

Even in an area as remote as the Amazon Basin, the present-day aerosol population is influenced by anthropogenic emissions, which are mostly from biomass burning. When the emission sources are large, as during the regional fire season, or located nearby, this influence can be readily identified. On the other hand, emissions arriving by long-range transport, especially from Africa, can have a significant influence on the aerosol population over Amazonia, even when there is little fire activity in the Amazon Basin (Prospero et al., 1981; Talbot et al., 1990; Swap et al., 1992; Formenti et al., 2001). For this reason, the CCN and CN concentrations summarized in Table 1 must always be considered as upper limits for the pristine values. The first CCN measurements in Amazonia were obtained during CLAIRE-98 by Roberts et al. (2001) at Balbina near Manaus. Subsequently, CCN and CN measurements were made as part of the LBA-EUSTACH and SMOCC programs (Andreae et al., 2002, 2004). In the course of a thorough investigation of the CCN counter used in the study of Roberts et al. (2001), it was found that the supersaturations in that study had been overestimated (Frank et al., 2007). Therefore, the values were corrected to a critical activation diameter of 85 nm at a supersaturation of 0.4%, corresponding to a $CCN_{0.4}$/CN ratio for the CLAIRE-98 data of 0.41. This critical diameter appears to be very robust for the clean Amazonian aerosol, and has been found independently in several studies (Rissler et al., 2004, D. Rose, personal communication, 2008). It has therefore also been used, where necessary, for the calculation of $CCN_{0.4}$ values from CN values in Table 1.

The results show surprisingly little difference between wet and dry season measurements in clean conditions. The CLAIRE-98, EUSTACH and AMAZE-08 measurements

during the full wet season (190±130, 155±65 and 138±94 cm^{-3}, respectively) are only moderately lower than the SMOCC-2002 values collected during the full dry season over the western Amazon (205±40 cm^{-3}). One can therefore conclude that the natural $CCN_{0.4}$ concentrations over Amazonia are centered around a value no greater than 180 cm^{-3}, with a range of about 80–250 cm^{-3}, and with only a modest seasonal range of about 40 cm^{-3}. Again, I emphasize that even these low values must contain some anthropogenic contamination from long-range transport.

During the burning season, pyrogenic aerosols from tens of thousands of fires overwhelm the natural aerosol population. It is not very meaningful to give average concentrations for this situation, as the actual concentration at any given time and place is dictated by the proximity to the fires and the meteorological conditions, and therefore varies over orders of magnitude from near-pristine values to those inside fresh plumes, which can be in the hundreds of thousands per cm^{-3}. Table 1 lists the ranges of typical concentrations observed during several campaigns in the smoky season. One finds that at sites remote from the fires, such as Balbina in northern Amazonas State, $CCN_{0.4}$ concentrations typically reach up to ca. 1000 cm^{-3}, while in the heavily impacted states of Mato Grosso and Rondonia, typical concentrations are in the range of 1000–4000 cm^{-3}.

Aerosol optical thickness measurements over the Amazon Basin from the AERONET network have been reviewed by Schafer et al. (2008). In Table 1 I give the averages from the cleanest 3 months (April–June in the northern part of the Amazon Basin, February–April in the southern part) to represent clean conditions, and the average of August and September in the southern part of the Amazon forest to represent the smoky period. The average values for the clean period fall near 0.09, while in the smoky period they range around 0.90, and thus show about the same factor-of-ten increase from clean to smoky conditions as the CCN concentrations. During some of the cleanest episodes, values down to 0.05 were observed. The Ångstrom exponent, å, during the clean periods is relatively low (0.7–1.1), indicating that coarse particles (primary biogenic material, but also some dust from long-range transport) contribute

significantly to AOT. The results from the Amazon forest are nearly identical to values observed at tropical and subtropical marine sites (cf. Table 2 and Fig. 1), such as Lanai (Hawaii) and San Nicolas Island (California), where AOT_{500} values of 0.08±0.03 have been observed, with å in the range of 0.6 to 1.3 (Holben et al., 2001).

During the smoky period, å increases to 1.7±0.1 as a result of the increased importance of the fine mode aerosol. The single scattering albedo of the smoke aerosol over the Amazon forest is 0.92±0.01, with excellent agreement between sunphotometer and in-situ measurements (Schmid et al., 2006; Schafer et al., 2008).

3.2 Remote temperate continental regions

Because the large expanses of temperate ecosystems fall into the same latitude belt as the regions with the highest density of anthropogenic emissions, it is very difficult to estimate pristine CCN concentrations and AOT over them. There is a surprisingly small number of data sets from remote regions in the temperate zone, and none where CCN and AOT measurements are truly collocated. Furthermore, the direct CCN measurements must either be taken from older studies, with sometimes uncertain measurement accuracy, or have to be deduced from size distributions. In spite of these problems, a surprisingly consistent picture emerges. In western and northern North America, remote sites tend to have $CCN_{0.4}$ concentrations ranging from about 90 in winter to ca. 280 in summer. Average AOT_{500} values typically fall in the 0.06–0.12 range. The northern European CCN concentrations tend to be somewhat higher, which is not altogether surprising considering the likely impact of residual air pollution in this region (Putaud et al., 2004; Van Dingenen et al., 2004). Unfortunately, no AOT data are available from this region.

An area of great potential interest for aerosol studies are the remote regions of Siberia, where the first measurements with modern aerosol size spectrometers have recently been reported (Heintzenberg et al., 2008). A preliminary analysis of a one year data set from the ZOTTO tall tower site near 60° N, 90° E shows an average $CCN_{0.4}$ concentration (calculated from CN>60 nm) of about 190 cm^{-3} during clean periods (de-

fined as those times when the aerosol absorption coefficient is <2 Mm^{-1}, corresponding to an equivalent black carbon concentration of about <0.2 $\mu g\,m^{-3}$). It is difficult to find corresponding AOT data, as the two nearest AERONET sites are in Krasnoyarsk and Tomsk, two highly industrialized cities with strong local pollution sources. The median and quartile ranges for AOT_{500} at these sites are 0.163(0.122, 0.219) and 0.138(0.094, 0.209), respectively. A better site to represent remote values in boreal Siberia may be Yakutsk, which has a much smaller population and little industry. The median AOT_{500} there is 0.081(0.053–0.120), comparable to many other remote continental sites, but still considerably higher than at Fort McMurray in northern Canada: 0.057(0.034, 0.089). The only report from the extratropical continental Southern Hemisphere is from a flight campaign in South Africa, where $CCN_{0.4}$ values of 137±63 were measured over the Highveld region on a clean day (Ross et al., 2003). On the same day, the AERONET site at Bethlehem, in the center of the Highveld, measured an AOT_{500} of 0.045±0.013.

In summary, the mean values of $CCN_{0.4}$ and AOT_{500} over extratropical remote sites are not distinctly different from those measured over Amazonia, even considering that some influence from long-range transport of pollution aerosol is unavoidable in these measurements. For example, more than half of the sulfate aerosol over remote British Columbia is from East Asian sources (van Donkelaar et al., 2008). This implies that pre-anthropogenic $CCN_{0.4}$ concentrations over most continental regions were below, maybe even well below, 200 cm^{-3}.

3.3 Remote marine regions

Remote marine regions, especially in the Southern Hemisphere, are usually considered the least polluted and most pristine parts of the atmosphere, but it must be remembered that anthropogenic pollution reaches even the remotest sites. This is, for example, readily seen at the Cape Grim background station, where aerosols from biomass burning in Southern Africa are readily detected during the fire season (Heintzenberg

and Bigg, 1990). Nevertheless, remote marine sites show the lowest number concentrations of aerosol particles and CCN worldwide. In the winter season, CCN concentrations in some areas drop down to a few tens per cm^3, as shown in Table 2 by data from the Southern Ocean (Cape Grim) and the Northeast Pacific (off Washington State), while in other areas, e.g. at the Northeast Atlantic coast (Mace Head) this seasonal cycle is less pronounced. Over biologically productive ocean regions, such as the tropical oceans and the temperate regions in summer, $CCN_{0.4}$ concentrations are typically near or above $100\,cm^{-3}$. This seasonal behavior is related to the biogenic production of marine aerosols, probably both via the emission of DMS and its oxidation to sulfate, and the release of primary biogenic particles (Charlson et al., 1987; Andreae and Rosenfeld, 2008). The reduced seasonality at some sites may be related to the effect of low levels of anthropogenic pollution, which can make a significant contribution to the very low aerosol concentrations present at remote oceanic sites in winter (Andreae et al., 1999, 2003; Reade et al., 2006).

Overall, one finds that CCN concentrations over the present-day remote oceans are on average about one-half of those over the present-day remote continents (Table 2), but with a very broad overlap (Fig. 1). In view of the fact that the sources of anthropogenic emissions are all located on land (with the exception of ship-stack emissions), it must be assumed that this ocean-land difference in CCN concentrations was substantially lower in pre-human times.

Remote ocean areas also show very low AOT values, in spite of the relatively high fraction of scattering associated with the seasalt aerosol (Quinn and Coffman, 1999). The datasets compiled in Table 2 yield an average AOT_{500} of 0.055±0.023, in good agreement with the "baseline" marine AOT_{550} of 0.06±0.01 given by Kaufman et al. (2005). Interestingly, the seasonality of AOT at Cape Grim is opposite to that of seasalt aerosol, which has its highest concentrations in the winter (Andreae et al., 1999). This argues against a dominant role of seasalt in controlling AOT, and suggests that the fine aerosol fraction may dominate both CCN concentration and AOT. The same conclusion was reached by Vallina et al. (2006) based on a statistical analysis of

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the CCN concentrations over the Southern Ocean and their controlling variables.

3.4 Polluted continental and marine regions

CCN concentrations and AOT values in polluted regions show a continuous increase from near-pristine values to extremely high levels in urban or biomass-burning regions (Table 2). In general, polluted marine regions tend to have lower values (range of study averages: $CCN_{0.4}$ 600–1700 cm^{-3}, AOT_{500} 0.15–0.39) than continental polluted areas ($CCN_{0.4}$ 370–9100 cm^{-3}, AOT_{500} 0.10–0.90) because they are usually more distant from sources. This can be also seen at some coastal sites, such as Amnyeon and Gosan (Korea), which experience both direct continental flow and inflow of polluted airmasses that have spent up to several days over the ocean. Thus, the $CCN_{0.4}$ concentrations in these airmasses, which in terms of airmass trajectories would be classified as marine, are in the range conventionally thought of as "continental", again putting in question the validity of this classification (Roberts et al., 2001). On the other hand, some of the continental sites in Europe and North America (e.g. the Hohenpeissenberg, New Hampshire and North Carolina sites) show relatively low CCN and AOT values, most likely as a result of the reductions in pollutant emissions over the last two decades. The highest values in Table 2 come from peri-urban regions in China, i.e. from locations just outside the urban areas of Guangzhou and Beijing. I have not included any urban measurements, because it is difficult to obtain representative measurements in such a highly variable environment, and because this analysis is mainly directed towards the regional to global scale.

3.5 Relationship between $CCN_{0.4}$ and AOT_{500}

The scatterplot between $CCN_{0.4}$ and AOT_{500} (Fig. 1) shows a surprisingly tight relationship, which can be fitted with a power law $AOT_{500}=0.0027\cdot[CCN_{0.4}]^{0.640}$ with a very high degree of correlation ($r^2=0.88$). While the deviations of individual studies from this trend are sometimes large (up to a factor of three), and obviously deviations for

single measurements at one place and time must be expected to be even greater, this does provide a basis for a parameterization of CCN concentrations in large-scale regional and global climate models. Note that in almost all cases the regression line goes through the error bars of the data. Figure 1 also highlights the broad overlap between remote marine and continental values.

Some further developments to this approach suggest themselves. Given that the CCN concentration is more closely tied to the finer fraction of the aerosol, AOT measurements at lower wavelengths might provide better correlations than the commonly used AOT_{500}, which was employed in this study. Alternatively, instead of AOT_{500}, one might examine correlations between CCN and the aerosol index AI, defined as the product of AOT and the Ångstrom exponent, thus providing another way of weighting the AOT measurement towards the fine mode. Finally, instead of the use of ground-based AOT measurements, one could examine the use of products based on satellite remote sensing, such as the fine mode AOT product from MODIS (Remer et al., 2005), especially once the remaining calibration issues in the MODIS products have been resolved. These investigations go beyond the scope of the present study, however, which has been designed as a first examination of large-scale relationships between potential proxies for the radiative and cloud microphysical forcings of anthropogenic aerosols.

Figure 2 shows the relationship between $CCN_{0.4}$ and CN concentrations. Again, there is a surprisingly good correlation, especially in view of the very different regimes from which the data are taken and the many different instruments by which they have been collected. The data in Table 2 suggest a fairly constant $CCN_{0.4}$/CN ratio of 0.36±0.14 (excluding the two very low values from Laramie, Wyoming), which has been plotted as a line in Fig. 2. This relationship reflects the relatively narrow range of hygroscopicity parameters and the convergent character of aerosol size distributions typical of many non-urban regions (Andreae and Rosenfeld, 2008).

4 Summary and conclusions

Analysis of published and unpublished data on AOT_{500} and CCN concentration shows that measurements from remote oceanic and continental regions fall into relatively narrow ranges. Remote marine $CCN_{0.4}$ concentrations are typically near or slightly above 100 cm^{-3} in biologically productive regions and seasons, and of the order of a few tens per cm^3 in winter. The average AOT_{500} over the remote oceans is 0.057±0.023, again with lower values in winter. Remote continental areas have, on average, almost twice as many CCN, and a mean AOT_{500} of 0.075±0.025 (Table 2). Some, or maybe even most of this difference, must be related to the closer proximity that even remote continental sites have to pollution sources, underscoring that the difference between marine and continental levels must have been smaller before the advent of anthropogenic pollution. CCN concentrations over polluted regions are on average about one order of magnitude greater than over their remote counterparts, while the AOT_{500} values over the polluted regions are about 5 times those over their remote equivalents (Table 2).

$CCN_{0.4}$ concentrations and AOT_{500} values show a surprising degree of correlation, which can be expressed as a power law (Note that regions dominated by desert dust have been excluded from this analysis). Given the difficulty of making direct CCN measurements, this relationship should be of great practical value in large-scale studies on the influence of the various direct and indirect aerosol effects on climate, as it provides an easily measured proxy for CCN concentrations. It also implies that the radiative and microphysical effects of aerosols on clouds, and therefore on climate and precipitation, are correlated and cannot vary independently of one another, at least not on larger scales. Various refinements to this analysis can be suggested, including the use of different AOT measurement wavelengths and spaceborne remote sensing.

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Table 1. $CCN_{0.4}$, CN, and AOT_{500} measurements from the Amazon Basin.

Campaign, Time, Season	$CCN_{0.4}$ (cm^{-3})	CN (cm^{-3})	AOT (500 nm)	Location	Reference
Clean conditions					
CLAIRE-98 Feb–Mar 1998, wet	190±130	460±320	0.089±0.020*	Balbina, Amazonas 01.92 S, 059.49 W	Roberts et al. (2001); Mar–Jun 2000–2001
LBA-EUSTACH Apr 1999, wet	155±65[a]	380±160	0.048	Rebio Jarú, Rondonia 10.08 S, 061.93 W	Guyon et al. (2003)
LBA-EUSTACH Jan–Mar, Nov 1999	230±100[a]	–	0.121±0.033*	Ji Paraná, Rondonia 10.88 S, 061.94 W	Williams et al. (2002)
CLAIRE-2001 Jul 2001, late wet	190±90	530±430	0.082±0.008*	Balbina, Amazonas 01.92 S, 059.49 W	Rissler et al. (2004)
SMOCC Oct 2002, late dry	205±40	500±100	–	NW of Cruzeiro do Sul, AM ~07 S, 073 W	Andreae et al. (2004)
AMAZE-08 Feb–Mar 2008, wet	138±94[a]	336±228	–	N of Manaus 02.60 S, 060.21 W	S. Gunthe, J. Schneider, unpubl.
AERONET 1993–2006	–	–	0.093±0.06	Amazon Basin forest	Schafer et al. (2008)
Smoky conditions					
LBA-EUSTACH Sep–Oct 1999, dry	1000–4000[a]	2000–8000	0.80±0.24	Rebio Jarú, Rondonia 10.08 S, 061.93 W	Guyon et al. (2003)
LBA-EUSTACH Sep–Oct 1999, dry	1300–7500[a]	2500–15 000	0.91	Fazenda Nossa Senhora, RO 10.76 S, 062.36 W	Artaxo et al. (2002)
LBA-EUSTACH Oct 1999, late dry	650–2000[a]	–	0.90*	Ji Paraná, Rondonia 10.88 S, 061.94 W	Williams et al. (2002)
CLAIRE-2001 Jul 2001, late wet	400–1000	800–2000	0.089±0.023*	Balbina, Amazonas 01.92 S, 059.49 W	Rissler et al. (2004)
SMOCC Sep–Oct 2002, dry	1000–4000	2000–8000	0.95*	Rondonia and Mato Grosso ~98–13 S, 056–064 W	Andreae et al. (2004)
AERONET 1993–2006	–	–	0.90±0.63	Southern Amazon Basin forest	Schafer et al. (2008)
Smoke plumes					
SMOCC Oct 2002, dry	10 000–22 000	20 000–44 000	–	Rondonia and Mato Grosso ~98–13 S, 056–064 W	Andreae et al. (2004)

* Data from the AERONET website. When the AERONET data period differs from the campaign period, it is indicated in the Reference column.

[a] $CCN_{0.4}$ calculated from CN using $CCN_{0.4}$/CN ratio (see text).

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Table 2. $CCN_{0.4}$, CN, and AOT_{500} measurements from remote and polluted, marine and continental environments. Data are presented as means and standard deviations (mmm±sss) or medians and quartile ranges [nnn (lll-uuu)]. "Æ:" identifies the AERONET site from which the AOT data are taken. Where the CCN and AOT data are from different locations, this is indicated in the Position column.

Location, Time	$CCN_{0.4}$ [cm^{-3}]	CN [cm^{-3}]	$CCN_{0.4}$/CN	AOT_{500}	Comment	Position	Reference
Clean conditions, marine							
Cape Grim summer winter	119±32 46±11	570±80 153±37	0.21 0.30	0.048±0.010 0.015±0.010	AOT data from CGO station	40.68 S, 144.69 E	Gras (1990); J. Gras, unpubl.; Wilson and Forgan (2002)
Southern Ocean, off Tasmania summer winter	90±20 27±10	266±150 210±190	0.34 0.13	0.048±0.010 0.015±0.010	aircraft, baseline AOT data from CGO station	~41–42 S, 144 E 40.68 S, 144.69 E	Yum and Hudson (2004)
S. Indian Ocean, Mar–Apr 2001	210±140	–	–	0.065 (0.048, 0.091)[b]	off S. Africa Æ: Reunion	20.88 S, 055.48 E	Ross et al. (2003)
S. Trop. Indian Oc. Feb/Mar 1999	150±20	361±31	0.42	0.058±0.035[b]	INDOEX Æ: Reunion	~3–8 S, ~072–074 E 20.88 S, 055.48 E	Hudson and Yum (2002)
Trop. S. Pacific Nov/Dec 2003/04	240±90	350±150	0.69	0.078 (0.060, 0.010)[b]	STRATUS 2003/04 Æ: Tahiti	~11–28 S, 71–90 W 17.58 S, 149.60 W	Tomlinson et al. (2007)
Trop. N. Pacific Jul–Aug 1990	80±50	180±110	0.44	0.065±0.026	near Hawaii Æ: Lanai, Jul/Aug 1997–2003	20.73 N, 156.92 W	Hudson (1993)
Temp. S. Pacific Nov–Dec 1995	108±44	330±70	0.33	–	ACE1	~41–51 S, 138–150 E	Hudson et al. (1998)
Temp. N. Pacific winter 1988–1990 summer 1989	23 78	252 594	0.09 0.13	–	off Washington	~47 N, 128 W	Hegg et al. (1991)
Temp. N. Pacific Jun–Jul 1987	55±35	170±80	0.32	– 0.072 (0.058, 0.096)	FIRE, below stratus Æ: San Nicolas Isl.	~31 N, 122 W 32.26 N, 119.49 W	Hudson and Frisbie (1991a)
Arctic Ocean May 1998	54±21 180±30	161±125 395±95	0.34 0.46	– –	below low cloud no low cloud	~76 N, 165 W	Yum and Hudson (2001)
Temp. North Atlantic Mace Head	120±50	–	–	–	little seasonality	53.33 N, 009.90 W	Jennings et al. (1998)
N. Atl., Mace Head summer winter	81±11 96±4 69±6	–	–	–	clean marine periods	53.33 N, 009.90 W	Reade et al. (2006)
Temp. N. Atlantic June 1992	155±50	380±150	0.41	–	ASTEX	~30–35 N, 18–25 W	Hudson and Xie (1999)
Temp. N. Atlantic 4 Jul 1997	190±50	910±160	0.21	0.095±0.018	ACE-2 Æ: Tenerife	~32–38 N, 011–013 W 28.03 N 016.63 W	Johnson et al. (2000)
Puerto Rico 13–14 Dec 2004 1–9 Jan 2005	108±54 101±42	290±100	0.37	0.065±0.020 0.060±0.016	Æ: Cape San Juan Dec 2005 1–9 Jan 2007	18.38 N, 065.62 W	Allan et al. (2008) G. Frank, unpubl.
Caribbean Sea Aug 1965	93	–	–	–	aircraft, BL ave.	~17 N, 066 W	Squires and Twomey (1966)
Tropical Atlantic Oct/Nov 1990	90±40	320±120	0.28	–	Atlantic transect	~10 N–30 S, 020–040 W	Schäfer et al. (1993)
Average	107±56	350±200	0.32±0.15	0.057±0.023			

[a] median

[b] AERONET level 1.5 data

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Table 2. Continued.

Location, Time	$CCN_{0.4}$ [cm^{-3}]	CN [cm^{-3}]	$CCN_{0.4}$/CN	AOT_{500}	Comment	Position	Reference
Clean conditions, continental							
Amazon Basin	185±120	440±100	0.41	0.082±0.020	clean conditions		from Table 1
Laramie, Wyoming Summer Winter	280±100 92±12	6800±3800 3180±1120	0.04 0.03	0.117 (0.081–0.179) 0.069 (0.051–0.106)	 Æ: Missoula, MT	~41 N, 104 W (CCN) 46.9 N, 114.08 W (Æ)	Delene and Deshler (2001)
Yukon Valley Feb 1972	90±10	–	–	–	–	~66 N, 148 W	Hoppel et al. (1973)
Colorado Plains Summer 1965	280	–	–	–	somewhat polluted	~40 N, 105 W	Squires and Twomey (1966)
Fort McMurray Canada	–	–	–	0.057 (0.034–0.089)	Æ: Fort McMurray	56.75 N, 111.48 W	
Pallas, Finland Apr 2000–Feb 2002	152±33	410	0.37	–	CN>80 nm, activated particles		Komppula et al. (2005)
Pallas, Finland winter summer	235	810 ~200 ~1200	0.29	–	CN>80 nm	67.97 N, 024.12 E	Lihavainen et al. (2003)
Hyytiälä, Finland 1996–2007	354[a]	2125	0.17	–	median, CN>65 nm	61.85 N, 024.30 E	M. Kulmala, unpubl.
Siberia, ZOTTO Sep 2006–Sep 2007	187 (109, 297)	283 (173, 446)	0.66	0.081 (0.053-0.120)	CN>60 nm, σ_s <2 Mm^{-1} Æ: Yakutsk	60.80 N, 089.35 E 61.66 N, 129.37 E (Æ)	W. Birmili, J. Heintzenberg, unpubl.
South Africa 1 Sep 2000	137±63	–	–	 0.045±0.013	S. Highveld, dry season Æ: Bethlehem	 28.25 S, 028.33 E (Æ)	Ross et al. (2003)
Average	200±90	2010±2370	0.38±0.18	0.075±0.025			

[a] median

[b] AERONET level 1.5 data

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Table 2. Continued.

Location, Time	$CCN_{0.4}$ [cm^{-3}]	CN [cm^{-3}]	$CCN_{0.4}$/CN	AOT_{500}	Comment	Position	Reference
Polluted conditions, marine							
North Atlantic Mace Head	630±380	–	–	–	polluted conditions	53.33 N, 009.90 W	Jennings et al. (1998)
NW Atlantic, off Nova Scotia Aug–Sep 1993	1170±700	4420±6160	0.26	0.15±0.06	NARE Æ: Keijmkuijk (Aug–Sep 1998–1999)	43.85 N, 066.12 W 44.38 N, 065.28 W	Liu et al. (1996)
Temp. N Atlantic June 1992	840±340	1390±550	0.60	–	ASTEX	~30–35 N, 18–25 W	Hudson and Xie (1999)
Temp. N Atlantic 16–18 Jul 1997 23–24 Jul 1997	660±200 830±130	2270±800 1710±200	0.29 0.49	0.32±0.09	ACE-2	~33–40 N, 011–014 W ~30–38 N, 011–013 W	Osborne et al. (2000) Wood et al. (2000)
Temp. NW Pacific Gosan, Korea	1570±500	3510±1790	0.45	0.28±0.02	ABC-EAREX	33.29 N, 126.16 W	Yum et al. (2007)
Temp. NW Pacific Anmyeon Isl., KR 1–22 May 2004	1670±390	3980±970	0.42	0.38±0.17	Æ: Amnyon	36.54 N, 126.33 E	Yum et al. (2005)
Indian Ocean, NH Feb/Mar 1999	1100±100	1810±40	0.61	0.39±0.17	INDOEX Æ: Kaashidoo	~0–4 N, ~072–074 E 4.96 N, 073.47 E	Hudson and Yum (2002)
Average	1060±400	2700±1200	0.44±0.14	0.30±0.10			
Polluted conditions, continental							
Mace Head, Ireland	370±70	–	–	–	polluted continental		Reade et al. (2006)
Amazon, smoky season	2500±1500	5000±3000	0.50	0.90±0.30	southern part of Amazon Basin	6–17 S, 45–70 W	From Table 1
Feldberg, near Frankfurt, Germany	2300±1000 1400±800	4650±1800 5700±4700	0.49 0.25	0.30±0.16 0.20±0.13	20 Jul–11 Aug 2004 22 Jun–6 Jul 2005 Æ: Mainz, summer 2004/06	freq. nucleation events 50.00 N, 008.30 E	U. Dusek, unpubl.
Hohenpeissenberg Germany, 1998–2000	1120±670	3130±2580	0.36	0.10±0.07	$CCN_{0.4}$=CN(>60 nm) 1999–2001, GAW	47.80 N, 011.12 E	A. Wiedensohler, unpubl. Birmili et al. (2003) GAW Brief Nr. 9
South Africa 1999–2001	740±460	–	–	0.17±0.15	wet and dry seasons	~−18–30 S, 25–32 E	Ross et al. (2003)
Reno, Nevada Dec 1988–May 1990	1310±580	8790±2000	0.15	–			Hudson and Frisbie (1991b)
New Hampshire Aug 2004	1090±350	~5000	0.22	0.24±0.21	rural site Æ: Billerica	42.53 N, 71.27 W	Medina et al. (2007)
North Carolina Duke Forest, Jul 2003	930	3400	0.27	0.38±0.16	Æ: Walker Branch	35.96 N, 84.29 W	Stroud et al. (2007)
Gosan, Korea 11 Mar–9 Apr 2005	2010±950	5600±3500	0.36	0.35±0.31	ABC-EAREX	33.29 N, 126.16 W	Yum et al. (2007)
Anmyeon Isl., KR 1–22 May 2004	3350±980	8310±1780	0.40	0.50±0.24	Æ: Amnyon	36.54 N, 126.33 E	Yum et al. (2005)
Beijing, 10 Aug–9 Sep 2006	7200±3000	16 200±8500	0.44	0.77±0.55	Yufa site Æ: Yufa	39.52 N, 116.33 E	D. Rose, unpubl.
Guangzhou Region NE monsoon Sep–Oct 2004	7300±3300	16 500±8800	0.44	0.80±0.38	CN>60 nm Xinken site Æ: Hong Kong Poly	22.60 N, 113.60 E 22.30 N, 114.18 E	A. Wiedensohler, unpubl.
Guangdong rural, SE monsoon, Jul 2006	9100±4800	18 700±8200	0.49	0.68±0.44	Æ: Backgarden site	23.49 N, 113.04 E	D. Rose, unpubl.
Average	2900±2800	8400±5500	0.36±0.12	0.45±0.27			

[a] median

[b] AERONET level 1.5 data

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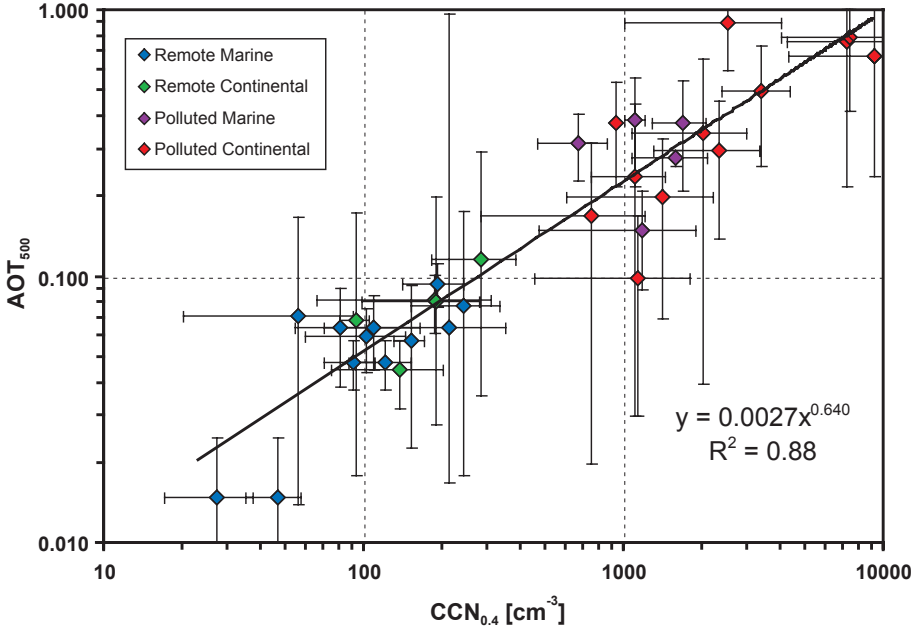

Fig. 1. Relationship between AOT_{500} and $CCN_{0.4}$ from investigations where these variables have been measured simultaneously, or where data from nearby sites at comparable times were available. The error bars reflect the variability of measurements within each study (standard deviations or quartiles).

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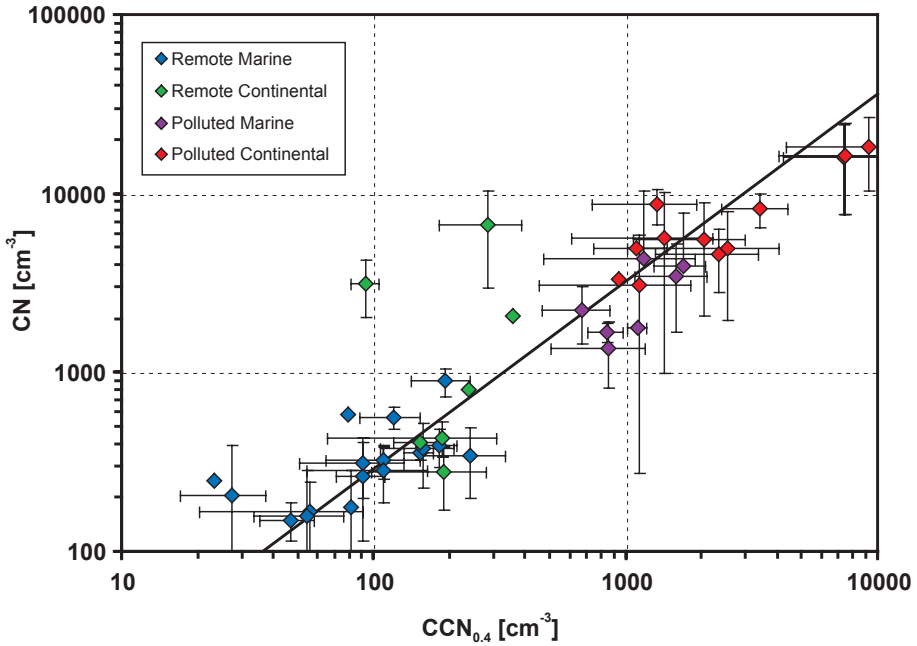

Fig. 2. Scatterplot of CN vs. $CCN_{0.4}$ based on the data from Table 2. The line represents the mean $CCN_{0.4}$/CN ratio of 0.36.

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